4. Air Monitoring

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Introduction

Air surveillance and air effluent monitoring are performed to evaluate compliance with local, state, and federal regulations, and to ensure that human health and the environment are protected from hazardous and radioactive air emissions. LLNL complies with local, state, and federal environmental air quality laws and DOE regulations including 40 CFR 61, the National Emissions Standards for Hazardous Air Pollutants (NESHAPs) section of the Clean Air Act; and DOE Orders 5400.1, *General Environmental Protection Program,* and 5400.5, *Radiation Protection of the Public and the Environment*. The *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. Department of Energy 1991) is the guidance for implementing DOE Orders 5400.1 and 5400.5. Other laws governing air quality include 22 CCR 67264.700 and 66265.710, Environmental and Compliance Monitoring, and the California Air Toxics "Hot Spots" Information and Assessment Act of 1987 (AB2588). In general, LLNL analyzes for most constituents at levels that are far below regulatory standards in order to determine any environmental impact.

LLNL monitors ambient air to determine if airborne radionuclides or hazardous materials are being released by Laboratory operations, what the concentrations are, and what the trends are in the LLNL environs. In addition, LLNL conducts air effluent monitoring at atmospheric discharge points of some facilities to determine the actual emissions from individual facilities and to confirm the operation of emission control systems. Air monitoring involves measurement of particles collected on filters or of vapor chemically trapped on a collection medium. Concentrations of various airborne radionuclides (including particles and tritiated water vapor) and beryllium are measured at the Livermore site, Site 300, at off-site locations throughout the Livermore Valley, and at an off-site location in Tracy. Point sources as well as diffuse, or nonpoint sources, are monitored to fulfill NESHAPs requirements.

Methods

For air surveillance monitoring, LLNL maintains eight continuously operating, high-volume, air particulate samplers on the Livermore site (**Figure 4-1**), ten in the Livermore Valley (**Figure 4-2**), eight at Site 300, and one in Tracy (**Figure 4-3**). One sampling location, LCCY, was removed in July of 1994 because of vandalism problems. The samplers are positioned to ensure reasonable probability that any significant concentration of particulate effluents from LLNL operations will be detected. Geographical details of particulate sampling locations are outlined in a written procedure in Appendix A of the *Environmental Monitoring Plan* (Tate et al. 1995).

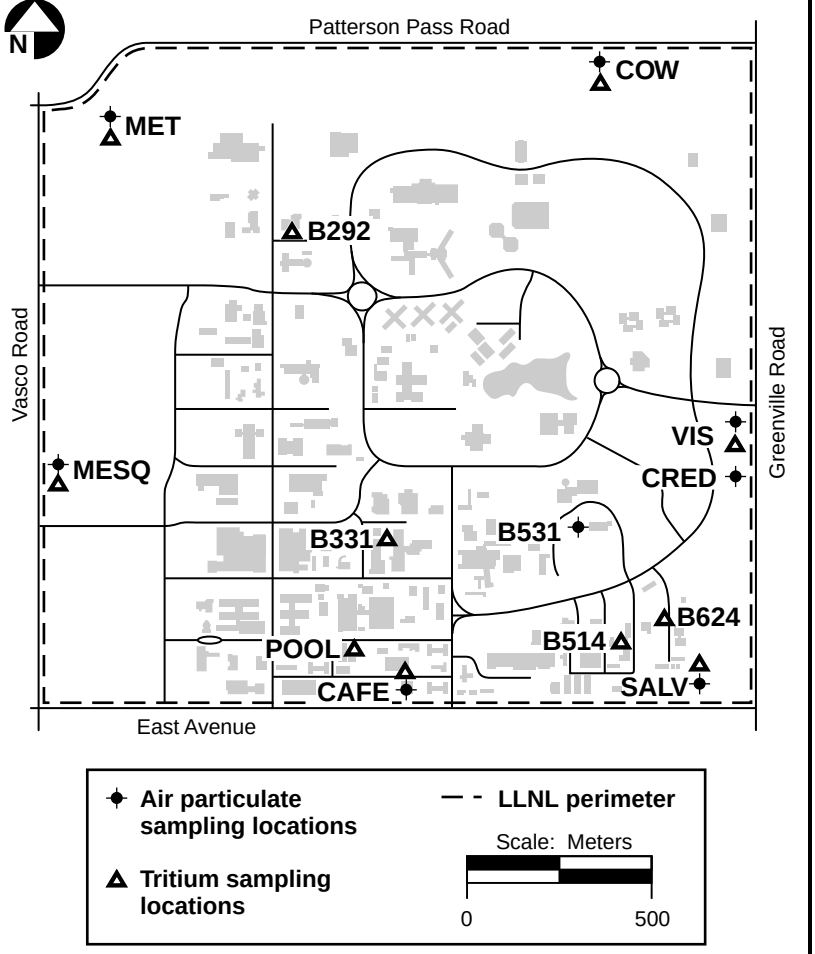

Figure 4-1. Air particulate and tritium sampling locations, Livermore site, 1994.

LLNL also maintains 11 continuously operating airborne tritium samplers on the Livermore site (**Figure 4-1**) and five samplers in the Livermore Valley (**Figure 4-2**). Four of the Livermore site locations (B331, B292, B514, and B624) monitor diffuse source emissions. The tritium sample locations are detailed in Appendix A of the *Environmental Monitoring Plan*.

Particulate filters are changed each week at all locations, and tritium samples are changed every two weeks. Replicate samples are processed to confirm the results obtained from the samplers. In addition, duplicate quality control (QC) samplers are operated for two months in parallel with the permanent sampler at a given site.

As outlined in the *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. Department of Energy 1991),

gross alpha and gross beta air filter results are used only as trend indicators; specific radionuclide analysis is done for plutonium, uranium, and all gamma emitters. All analytical results are reported as a measured concentration per volume of air, or at the minimum detection limit (MDL) when no activity is detected. In all cases, the MDL is more than adequate for demonstrating compliance with the pertinent regulatory requirements for radionuclides that may be or are present in the air sample. Particle size distributions are not determined because the estimated effective dose equivalent to the maximally exposed individual is well below the 0.01 mSv (1 mrem) allowable limit. Further details of the surveillance monitoring methods are included in Volume 2, Chapter 4.

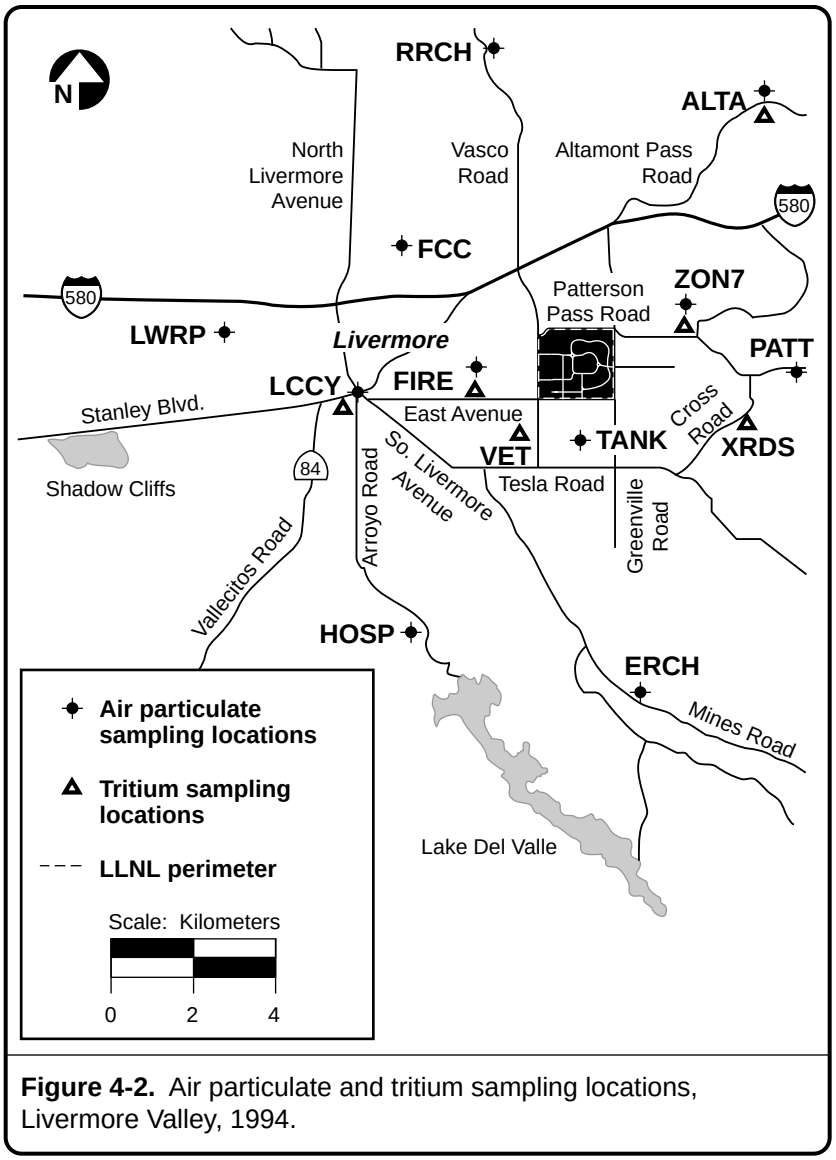

Figure 4-2. Air particulate and tritium sampling locations, Livermore Valley, 1994.

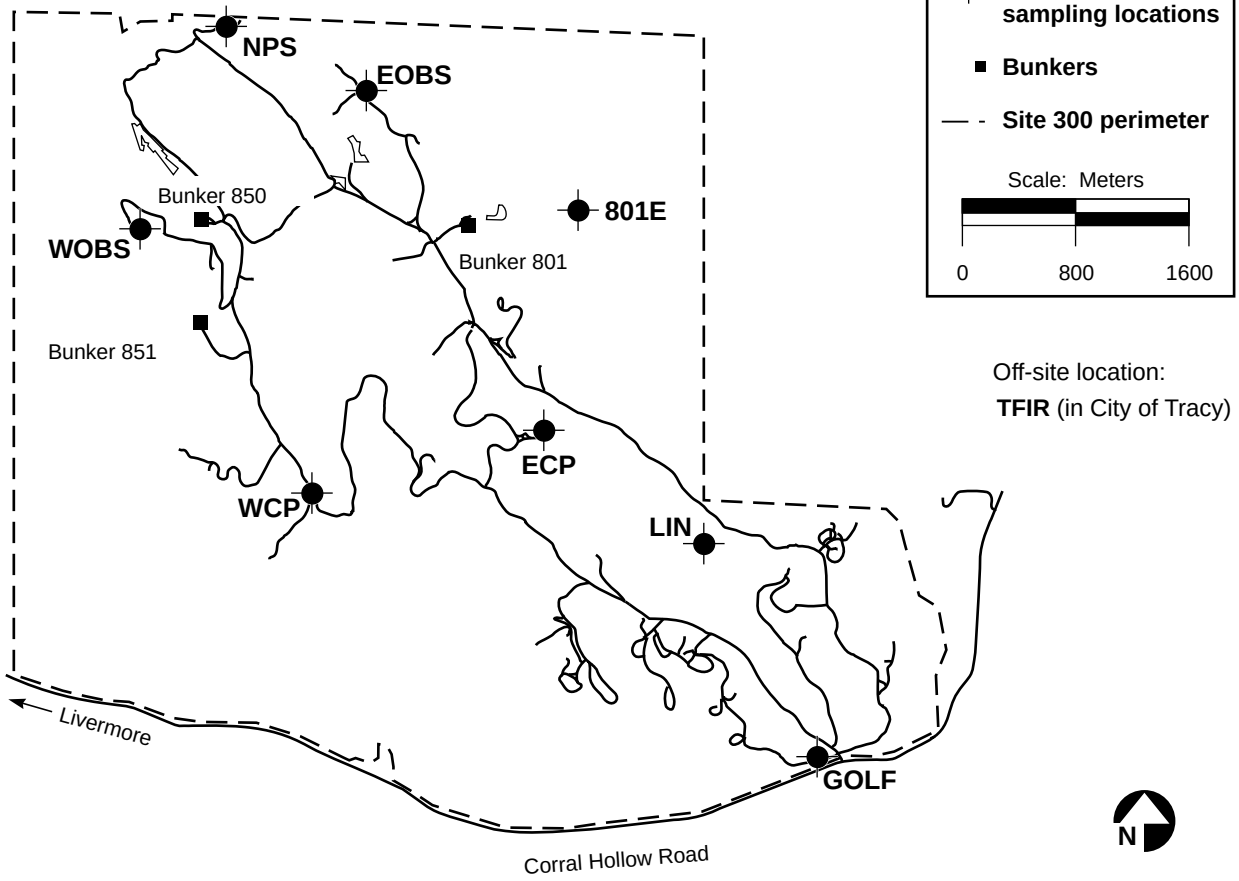

Figure 4-3. Air particulate sampling locations, Site 300, 1994.

For air effluent monitoring, LLNL maintains 92 radionuclide sampling systems on air exhausts at eight facilities at the Livermore site. These systems are listed in **Table 4-1** along with the analytes of interest, the type of sampler, and the number of samplers and discharge points monitored. Sampling for particles containing radionuclides is conducted in seven of the facilities; sampling for tritium is conducted in one facility. All sampling systems operate continuously. Samples are changed weekly or biweekly depending on the facility. Air samples for particulate emissions are extracted downstream of high efficiency particulate air (HEPA) filters and prior to the discharge point to the atmosphere. Particles in the extracted air are collected on sample filters and analyzed for gross alpha and beta activity. Tritium is collected using molecular sieves. In addition to sample collection for environmental reporting, some facilities have real-time monitors at discharge points to provide faster notification in the event of a release of radioactivity. Further details of LLNL air effluent sampling systems are included in Chapter 4 of the *Environmental Monitoring Plan*.

Table 4-1. Air effluent sampling locations and systems.

Building	Facility	Analytes	Sample Type	Number of Samplers	Number of Discharge Points
175	Mars	Gross α,β on particles	Filters	6	6
231	Vault	Gross α,β on particles	Filter	1	1
251	Heavy elements				
	Unhardened area	Gross α,β on particles	Filters	44	55[a]
	Hardened area	Gross α,β on particles	CAM[b]	4	4
331	Tritium	Tritium	Ionization chamber[b]	4	4
		Gaseous tritium/tritiated water vapor	Molecular sieves	4	2
332	Plutonium	Gross α,β on particles	CAM[b]	12	11
		Gross α,β on particles	Filters	12	11
419	Decontamination	Gross α,β on particles	Filters	2	2
490	Laser isotope separation	Gross α,β on particles	Filters	4	4
491	Laser isotope separation	Gross α,β on particles	Filters	1	1

Note: "CAM" denotes Eberline continuous air monitors.

a Alternate blower system measured by the same sampler.

b Alarmed systems.

The need for continuous air effluent monitoring at other air discharge points that can potentially release radionuclides to the atmosphere is evaluated according to the NESHAPs regulations. The evaluation is based on estimated releases using radionuclide inventories specific to individual discharge points and does not take into account reduction by emission control systems (according to the regulations). As reported in the *LLNL NESHAPs 1994 Annual Report* (Surano et al. 1995), no additional locations were identified that require continuous monitoring. In fact, many of the existing sampling systems now in place (**Table 4-1)** are not required; however, LLNL has continued to operate these systems as a best-management practice.

Results

This section discusses the air monitoring results at the Livermore site and at Site 300.

Livermore Site

Airborne Radioactivity

Table 4-2 shows the monthly gross alpha and gross beta detection frequency, median, interquartile range (IQR), and maximum for all Livermore Valley, Livermore-site perimeter, and Site 300 sampling locations. (See Volume 2, Tables 4-1 and 4-2, for a weekly summary of Livermore Valley and Livermore-site perimeter gross alpha and gross beta concentrations in air.) Negative values are not considered detections. The monthly median gross alpha and gross beta concentrations in air are plotted in **Figures 4-4** and **4-5**, respectively. The gross beta results seem to be much more variable and higher during the fall and winter. This apparent seasonal pattern is similar to 1992 and 1993 data. The values reported for gross alpha and gross beta activities are similar to those observed in previous years and show no significant differences between samples taken at the Livermore-site perimeter, Livermore Valley, and Site 300. Most of the gross alpha determinations were at or near the detection limit of the method. Typical gross alpha activity for the Livermore Valley is -4.7×10^{-12} Bq/mL (-1.3×10^{-22} Ci/mL) and for the Livermore-site perimeter is -3.9×10^{-12} Bq/mL (-1.1×10^{-22} Ci/mL). Approximately 56% of the gross alpha values are negative. The negative values occur because the background of the devices used to analyze the filters is higher than the amount of activity on the filters. Typical gross beta activity for the Livermore Valley is 4.1×10^{-10} Bq/mL (1.1×10^{-20} Ci/mL) and 4.5×10^{-10} Bq/mL (1.2×10^{-20} Ci/mL) for the Livermore-site perimeter. The primary sources of observed alpha and beta activity are the naturally occurring radioisotopes of uranium and thorium and their decay products.

Table 4-3 shows a summary of gamma-emitting radionuclide concentrations in air that contribute to the activity in the Livermore-site perimeter samples. (See Volume 2, Table 4-4 for monthly gamma data.) Of the nuclides tabulated, ^{7}Be, ^{40}K, ^{226}Ra, ^{228}Ra, and ^{228}Th occur naturally. The primary source of ^{137}Cs is long-term global fallout and fallout resuspension.

In addition to providing baseline data on global fallout, analysis of these radionuclides enables LLNL to monitor the containment of the small inventories of mixed fission products and radiochemical tracers used at LLNL. The Derived Concentration Guides (DCGs) for these radionuclides are also shown in **Table 4-3**. For air, DCGs specify the concentrations of radionuclides that could be inhaled continuously 365 days a year without exceeding the DOE primary radiation protection standard for the public, which is 1 mSv/y (100 mrem/y) effective dose equivalent (DOE Order 5400.5). (Chapter 12 on Radiological Dose Assessment provides an explanation of this and other units of dose.) Finally, the fraction of the DCGs is presented. These values demonstrate the low levels of gamma activity present in air at the Livermore-site perimeter.

Table 4-2. Gross alpha and gross beta (Bq/mL) in air particulate samples summarized by month, 1994.

	Detection Frequency	Monthly Median	Interquartile Range	Maximum
Livermore Valley				
Gross alpha				
Jan	21/38	2.8×10^{-12}	2.6×10^{-11}	8.8×10^{-11}
Feb	36/43	4.7×10^{-11}	6.8×10^{-11}	1.8×10^{-10}
Mar	22/51	-1.0×10^{-11}	3.2×10^{-11}	1.0×10^{-10}
Apr	17/41	-6.3×10^{-12}	1.4×10^{-11}	1.0×10^{-10}
May	18/39	-6.1×10^{-12}	3.4×10^{-11}	1.3×10^{-10}
Jun	13/54	-2.9×10^{-11}	—[a]	7.7×10^{-11}
Jul	15/38	-2.2×10^{-11}	2.5×10^{-11}	1.1×10^{-10}
Aug	17/39	-4.9×10^{-12}	1.9×10^{-11}	1.1×10^{-10}
Sep	29/49	1.4×10^{-11}	2.5×10^{-11}	1.3×10^{-10}
Oct	11/37	-2.4×10^{-11}	2.3×10^{-11}	7.4×10^{-11}
Nov	19/36	2.2×10^{-12}	4.1×10^{-11}	8.9×10^{-11}
Dec	18/46	-1.4×10^{-11}	2.6×10^{-11}	1.0×10^{-10}
Gross beta				
Jan	38/38	1.1×10^{-9}	1.1×10^{-9}	2.7×10^{-9}
Feb	43/43	4.4×10^{-10}	6.0×10^{-10}	2.2×10^{-9}
Mar	51/51	4.4×10^{-10}	1.6×10^{-10}	6.6×10^{-10}
Apr	41/41	2.7×10^{-10}	1.2×10^{-10}	6.4×10^{-10}
May	39/39	2.5×10^{-10}	1.7×10^{-10}	4.6×10^{-10}
Jun	54/54	3.1×10^{-10}	1.9×10^{-10}	5.4×10^{-10}
Jul	38/38	2.6×10^{-10}	2.0×10^{-10}	6.7×10^{-10}
Aug	39/39	3.5×10^{-10}	1.1×10^{-10}	5.4×10^{-10}
Sep	49/49	8.0×10^{-10}	5.9×10^{-10}	1.2×10^{-9}
Oct	37/37	7.5×10^{-10}	6.6×10^{-10}	3.1×10^{-9}
Nov	36/36	3.7×10^{-10}	2.3×10^{-10}	9.7×10^{-10}
Dec	46/46	5.7×10^{-10}	3.5×10^{-10}	1.2×10^{-9}
Livermore Perimeter				
Gross alpha				
Jan	9/22	-3.3×10^{-11}	1.0×10^{-11}	1.0×10^{-10}
Feb	16/21	3.6×10^{-11}	8.4×10^{-11}	1.1×10^{-10}
Mar	12/28	-6.7×10^{-12}	3.6×10^{-11}	1.3×10^{-10}
Apr	9/24	-1.3×10^{-11}	1.3×10^{-11}	1.3×10^{-10}
May	13/24	5.4×10^{-12}	2.5×10^{-11}	9.5×10^{-11}
Jun	11/27	-1.7×10^{-11}	2.9×10^{-11}	7.4×10^{-11}
Jul	10/22	-2.8×10^{-12}	4.2×10^{-11}	1.0×10^{-10}
Aug	15/24	5.3×10^{-12}	3.4×10^{-11}	1.4×10^{-10}
Sep	14/30	-2.1×10^{-12}	3.6×10^{-11}	1.1×10^{-10}
Oct	9/23	-1.8×10^{-11}	1.8×10^{-11}	4.8×10^{-11}
Nov	14/24	7.3×10^{-12}	3.8×10^{-11}	1.0×10^{-10}
Dec	9/29	-1.5×10^{-11}	1.2×10^{-11}	6.6×10^{-11}

Table 4-2. Gross alpha and gross beta (Bq/mL) in air particulate samples summarized by month, 1994 (concluded).

	Detection Frequency	Monthly Median	Interquartile Range	Maximum
Livermore Perimeter (continued)				
Gross beta				
Jan	22/22	1.6×10^{-9}	1.3×10^{-9}	3.7×10^{-9}
Feb	21/21	4.7×10^{-10}	4.4×10^{-10}	1.8×10^{-9}
Mar	28/28	4.7×10^{-10}	1.3×10^{-10}	6.7×10^{-10}
Apr	24/24	3.0×10^{-10}	1.4×10^{-10}	5.3×10^{-10}
May	23/24	2.5×10^{-10}	2.0×10^{-10}	5.4×10^{-10}
Jun	27/27	2.9×10^{-10}	1.4×10^{-10}	6.3×10^{-10}
Jul	22/22	3.2×10^{-10}	1.9×10^{-10}	6.7×10^{-10}
Aug	24/24	4.0×10^{-10}	2.0×10^{-10}	6.8×10^{-10}
Sep	30/30	7.5×10^{-10}	4.9×10^{-10}	1.4×10^{-9}
Oct	23/23	5.8×10^{-10}	9.1×10^{-10}	2.1×10^{-9}
Nov	24/24	3.6×10^{-10}	3.1×10^{-10}	7.2×10^{-10}
Dec	29/29	6.5×10^{-10}	4.3×10^{-10}	1.1×10^{-9}
Site 300				
Gross Alpha				
Jan	19/34	1.2×10^{-11}	4.3×10^{-11}	8.6×10^{-11}
Feb	27/36	1.8×10^{-11}	4.8×10^{-11}	1.5×10^{-10}
Mar	10/43	-2.7×10^{-11}	—[a]	1.3×10^{-10}
Apr	19/36	7.0×10^{-13}	1.8×10^{-11}	6.6×10^{-11}
May	13/36	-7.6×10^{-12}	2.4×10^{-11}	6.6×10^{-11}
Jun	10/44	-2.6×10^{-11}	—[a]	9.6×10^{-11}
Jul	7/35	-4.7×10^{-11}	—[a]	6.7×10^{-11}
Aug	21/35	9.3×10^{-12}	3.1×10^{-11}	1.0×10^{-10}
Sep	18/44	-1.2×10^{-12}	2.8×10^{-11}	1.3×10^{-10}
Oct	13/36	-1.3×10^{-11}	9.9×10^{-12}	7.5×10^{-11}
Nov	10/36	-1.4×10^{-11}	1.9×10^{-12}	5.8×10^{-11}
Dec	14/40	-1.8×10^{-11}	1.2×10^{-11}	1.3×10^{-10}
Gross Beta				
Jan	34/34	9.7×10^{-10}	1.2×10^{-9}	3.1×10^{-9}
Feb	36/36	3.8×10^{-10}	4.9×10^{-10}	2.3×10^{-9}
Mar	43/43	3.6×10^{-10}	1.4×10^{-10}	6.8×10^{-10}
Apr	36/36	2.6×10^{-10}	1.8×10^{-10}	6.3×10^{-10}
May	36/36	2.9×10^{-10}	2.5×10^{-10}	5.4×10^{-10}
Jun	44/44	3.6×10^{-10}	1.4×10^{-10}	6.7×10^{-10}
Jul	35/35	4.4×10^{-10}	1.7×10^{-10}	7.0×10^{-10}
Aug	35/35	4.5×10^{-10}	1.9×10^{-10}	8.2×10^{-10}
Sep	44/44	8.0×10^{-10}	5.2×10^{-10}	1.2×10^{-9}
Oct	36/36	7.8×10^{-10}	8.2×10^{-10}	2.0×10^{-9}
Nov	36/36	3.9×10^{-10}	3.3×10^{-10}	1.1×10^{-9}
Dec	40/40	5.0×10^{-10}	3.6×10^{-10}	1.5×10^{-9}

[a] Interquartile range not calculated. See Chapter 14, Quality Assurance.

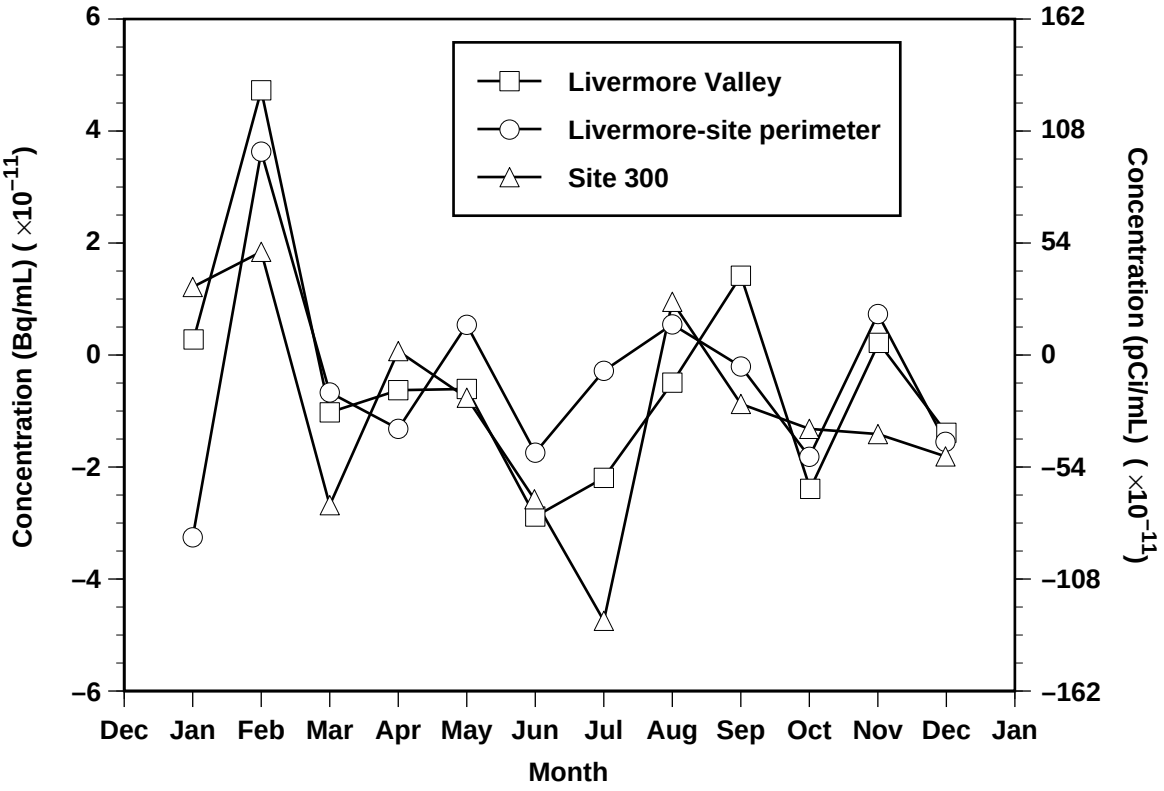

Figure 4-4. Monthly median gross alpha concentrations on air filters from Livermore Valley, Livermore-site perimeter, and Site 300 sampling locations.

Table 4-4 shows the detection frequency, median, IQR, maximum, and fraction of DCG for concentration of plutonium on air filter samples collected in the Livermore Valley. (See Volume 2, Table 4-6 for monthly data.) The highest off-site median concentration of ^{239}Pu occurred at the Livermore Water Reclamation Plant (LWRP). Soils near the LWRP contain some detectable plutonium, principally resulting from sludge-spreading operations following an estimated 1.2×10^9 Bq (32 mCi) release to the sewer in 1967 (see Chapter 9, Soil and Sediment Monitoring). Resuspension of these soils probably accounts for the slightly higher average ^{239}Pu in air concentrations observed. However, the median observed value is <0.0001 of the DCG.

Table 4-4 also shows the concentrations of airborne ^{239}Pu on air filter samples from the Livermore-site perimeter. (See Volume 2, Table 4-7 for monthly data.) The August sample for location CAFE, which is on the south perimeter of the Livermore site, registered the highest concentrations of ^{239}Pu in air of all perimeter sampling locations. The concentration reported for this sample,

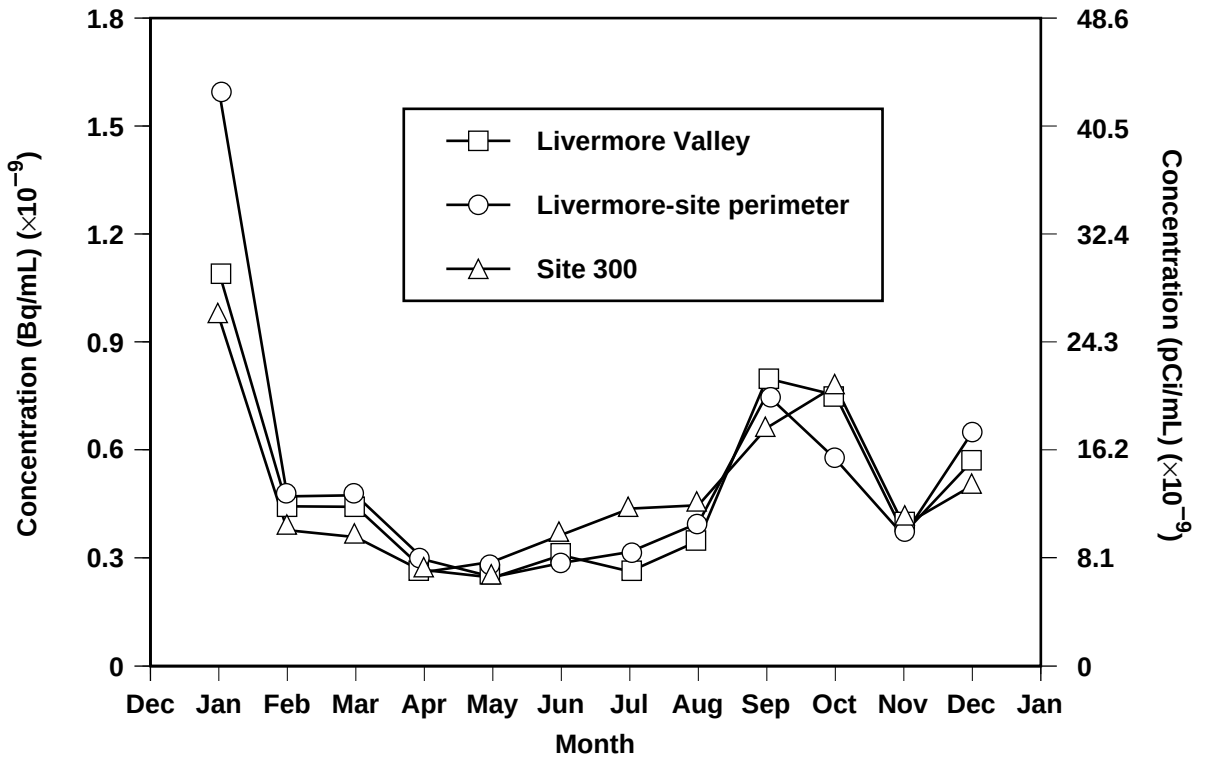

Figure 4-5. Monthly median gross beta concentrations on air filters from Livermore Valley, Livermore-site perimeter, and Site 300 sampling locations.

3.4×10^{-13} Bq / mL (9.1×10^{-24} Ci / mL), represents 0.0005 of the DCG. The annual median concentration of ^{239}Pu at location CAFE was 3.4×10^{-14} Bq / mL (9.2×10^{-25} Ci / mL). No other statistically significant differences between locations or samples were noted, and the overall ^{239}Pu levels were similar to those reported in 1993.

Figure 4-6 shows the annual median concentrations of ^{239}Pu for locations SALV (on-site) and FCC (off-site) from 1982 to 1994. Location FCC represents a typical upwind background location, and SALV represents the perimeter location having the highest annual average for most of this 13-year period. The higher values in the past at SALV may be attributed to historical activities at LLNL.

In June 1991, two air particulate sampling locations (B531 and CRED) were added as part of a special study to investigate the somewhat elevated levels of plutonium in air and surface soil in the southeast quadrant of the Livermore site (see Chapter 9, Soil and Sediment Monitoring, for general background on this

Table 4-3. Gamma activity on air filters, Livermore-site perimeter and Site 300, 1994.

	(10^{-9} Bq/mL)	(10^{-12} Bq/mL)					
	^{7}Be	^{40}K	^{137}Cs	^{22}Na	^{226}Ra	^{228}Ra	^{228}Th
Livermore Perimeter							
Median	4.61	<7.07	<0.24	<0.26	<0.47	<1.07	<0.64
Interquartile range	1.45	<24.62	—[a]	—[a]	—[a]	—[a]	—[a]
Maximum	6.14	27.08	0.48	0.63	1.85	3.52	1.39
Median fraction of DCG[b]	3.1×10^{-6}	$<2.1 \times 10^{-7}$	$<1.6 \times 10^{-8}$	$<7.0 \times 10^{-9}$	$<1.3 \times 10^{-5}$	$<9.7 \times 10^{-6}$	$<4.3 \times 10^{-4}$
Site 300							
Median	5.11	<4.37	<0.15	<0.48	<0.33	<0.59	<0.41
Interquartile range	2.57	—[a]	—[a]	<0.67	—[a]	—[a]	—[a]
Maximum	7.77	14.17	0.51	1.00	1.27	1.86	1.22
Median fraction of DCG[b]	3.4×10^{-6}	$<1.3 \times 10^{-7}$	$<1.0 \times 10^{-8}$	$<1.3 \times 10^{-8}$	$<9.0 \times 10^{-6}$	$<5.3 \times 10^{-6}$	$<2.8 \times 10^{-4}$
DCG[b] (Bq/mL)	1.5×10^{-3}	3.3×10^{-5}	1.5×10^{-5}	3.7×10^{-5}	3.7×10^{-8}	1.1×10^{-7}	1.5×10^{-9}

a No measure of dispersion calculated. See Chapter 14, Quality Assurance.

b Derived Concentration Guide.

study). These sampling locations are now part of our routine monitoring network and provide data for diffuse source dose assessments. **Table 4-4** shows the median concentrations of airborne ^{239}Pu at these two locations. (See Volume 2, Table 4-8 for monthly data.) The median concentration of 1.7×10^{-13} Bq / mL (4.5×10^{-24} Ci / mL) at location B531 is higher than the median concentration for any of the other air particulate sampling locations but is still only 0.0002 of the DCG.

The median ^{235}U and ^{238}U concentrations in air samples from the Livermore-site perimeter are shown in **Table 4-5**. (See Volume 2, Table 4-10 for monthly data.) The maximum measured concentrations of ^{238}U are less than 0.0004 of the DCG (DOE Order 5400.5). All ^{235}U / ^{238}U median ratios are as expected for naturally occurring uranium; however, monthly data in Volume 2 shows some unexpected ^{235}U / ^{238}U ratios for natural uranium around the Livermore-site perimeter. The cause of these apparent depleted and enriched uranium ratios is not known, but they have occurred sporadically in the past.

Table 4-4. Plutonium activity on air filters (in 10^{-15} Bq/mL), 1994.

Sampling Location[a]	Detection Frequency	Median	Interquartile Range	Maximum	Median Fraction of DCG[b]
Livermore Valley					
TANK	11/12	3.53	6.05	12.91	4.8×10^{-6}
ZON7	10/12	7.38	9.90	16.61	1.0×10^{-5}
FCC	7/12	1.33	7.15	64.75	1.8×10^{-6}
HOSP	8/12	3.34	7.67	16.84	4.5×10^{-6}
LWRP	10/12	11.08	33.1	65.86	1.5×10^{-5}
FIRE	8/12	4.22	10.46	29.12	5.7×10^{-6}
TFIR	9/12	5.92	8.58	19.31	8.0×10^{-6}
ALTA	7/12	2.13	6.08	43.66	2.9×10^{-6}
ERCH	10/12	6.64	10.71	34.11	9.0×10^{-6}
LCCY	3/6	2.93	7.66	18.50	4.0×10^{-6}
RRCH	8/12	3.48	9.72	11.84	4.7×10^{-6}
PATT	7/12	4.02	7.97	41.81	5.4×10^{-6}
Livermore-Site Perimeter					
SALV	12/12	18.59	7.38	37.37	2.5×10^{-5}
MESQ	12/12	23.07	6.56	63.64	3.1×10^{-5}
CAFE	12/12	34.08	10.78	338.9	4.6×10^{-5}
MET	12/12	21.70	14.74	50.69	2.9×10^{-5}
VIS	12/12	22.31	14.34	62.16	3.0×10^{-5}
COW	12/12	23.27	10.84	48.47	3.1×10^{-5}
Diffuse On-Site Sources					
B531	12/12	166.0	346.7	521.7	2.2×10^{-4}
CRED	10/12	10.27	13.79	38.11	1.4×10^{-5}
Site 300	12/12	4.31	3.74	11.80	5.8×10^{-6}

a See Figures 4-1, 4-2, and 4-3 for sampling locations. Location TFIR is in Tracy.

b DCG = 7.4×10^{-10} Bq/mL for ^{239}Pu activity in air (2×10^{-14} μCi/mL).

Table 4-5. Uranium activity on air filters, 1994.

Sampling Location[a]	Uranium-238[b] [10^{-5} μg/m^3]	Uranium-235[c] [10^{-7} μg/m^3]	Uranium-235/238[d] [10^{-3}]
Livermore Perimeter			
SALV			
Median	3.50	3.36	7.34
Interquartile range	3.63	3.01	1.17
Maximum	9.78	7.02	
Median fraction of DCG	1.2×10^{-4}	7.1×10^{-6}	
MESQ			
Median	4.32	3.17	7.15
Interquartile range	2.74	2.28	0.46
Maximum	9.00	6.93	
Median fraction of DCG	1.4×10^{-4}	6.7×10^{-6}	
CAFE			
Median	5.45	3.88	7.13
Interquartile range	3.42	2.68	0.27
Maximum	11.60	8.39	
Median fraction of DCG	1.8×10^{-4}	8.2×10^{-6}	
MET			
Median	3.65	2.60	7.16
Interquartile range	3.29	2.55	0.54
Maximum	8.86	7.03	
Median fraction of DCG	1.2×10^{-4}	5.5×10^{-6}	
VIS			
Median	3.28	2.39	7.30
Interquartile range	1.62	1.18	0.23
Maximum	7.96	5.64	
Median fraction of DCG	1.1×10^{-4}	5.1×10^{-6}	
COW			
Median	5.06	3.72	7.22
Interquartile range	3.96	2.80	0.34
Maximum	10.80	7.64	
Median fraction of DCG	1.7×10^{-4}	7.9×10^{-6}	
Site 300			
Site 300 composite			
Median	4.84	2.90	6.72
Interquartile range	4.61	2.49	1.00
Maximum	50.0	21.0	
Median fraction of DCG	1.6×10^{-4}	6.2×10^{-6}	

a See Figures 4-1 and 4-3 for sampling locations.

b DCG = 0.3 μg/m^3 for ^{238}U activity in air.

c DCG = 0.047 μg/m^3 for ^{235}U activity in air.

d Naturally occurring uranium has a ^{235}U/^{238}U ratio of 7.1×10^{-3}. Maximum not computed for ^{235}U/^{238}U ratio.

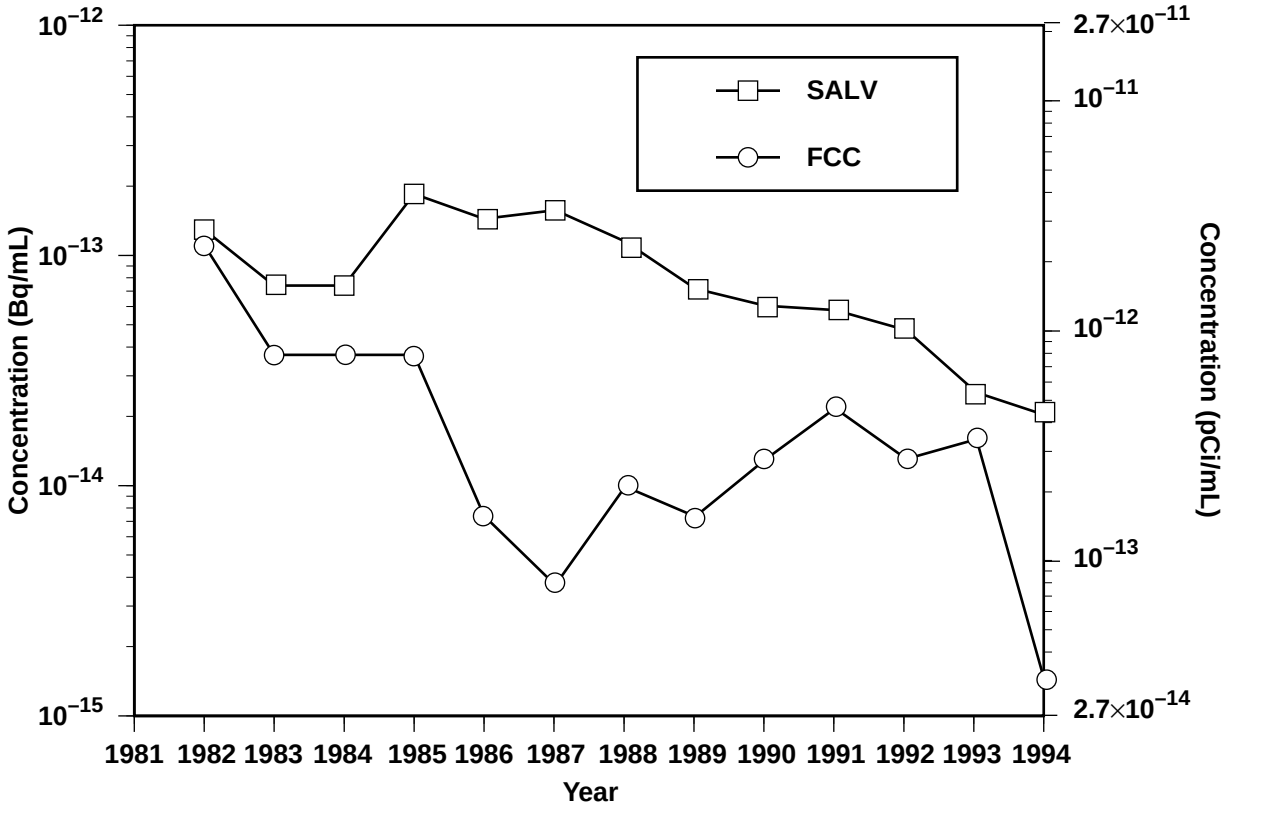

Figure 4-6. Median plutonium concentrations on air filters at two locations, SALV and FCC, 1982 to 1994.

Table 4-6 shows the median concentrations of tritiated water vapor for the Livermore Valley sampling locations. (See Volume 2, Table 4-12 for biweekly data for each location.) The highest annual median concentration was observed at location ZON7. At approximately 4.4×10^{-8} Bq / mL (1.2×10^{-18} Ci / mL), this concentration represents 0.00001 of the DCG. The highest biweekly concentration was observed in December at VET. If it were a yearly average, this concentration, 1.4×10^{-7} Bq / mL (3.8×10^{-18} Ci / mL), would be 0.00004 of the DCG. The 1994 tritium values generally are similar to those reported last year.

Table 4-6 also shows the median concentrations of tritiated water vapor that were observed at the Livermore-site perimeter sampling locations. (See Volume 2, Table 4-13 for biweekly data.) The highest annual median concentration was observed at location POOL, which was 1.5×10^{-7} Bq / mL (4.1×10^{-18} Ci / mL), or 0.00004 of the DCG.

Diffuse sources of tritium on the Livermore site are monitored at air tritium sampling locations B331, B292, B514, and B624. **Table 4-6** shows the median concentrations of tritiated water vapor for these sampling locations. (See

Table 4-6. Tritium in air (in 10^{-9} Bq/mL), 1994.

Sampling Location[a]	Detection Frequency	Median	IQR[b]	Maximum	Median Fraction of DCG[c]	Median Dose (mSv)[d]
Livermore Valley						
ZON7	25/26	44.2	21.3	109.9	1.2×10^{-5}	9.5×10^{-6}
ALTA	15/24	<15.4	—[e]	30.8	$<4.2 \times 10^{-6}$	3.3×10^{-6}
LCCY	7/8	16.3	3.6	32.1	4.4×10^{-6}	3.5×10^{-6}
FIRE	11/24	<15.2	<23.3	55.5	$<4.1 \times 10^{-6}$	3.3×10^{-6}
XRDS	18/25	<18.5	<34.7	79.9	$<5.0 \times 10^{-6}$	4.0×10^{-6}
VET	17/24	35.1	<58.7	142.5	9.5×10^{-6}	7.5×10^{-6}
Livermore Perimeter						
SALV	24/24	116.7	100.0	283.1	3.2×10^{-5}	2.5×10^{-5}
MESQ	19/25	43.3	51.5	115.4	1.2×10^{-5}	9.3×10^{-6}
CAFE	26/26	91.9	78.3	175.0	2.5×10^{-5}	2.0×10^{-5}
MET	21/26	29.6	28.3	105.5	8.0×10^{-6}	6.4×10^{-6}
VIS	26/26	91.4	54.1	178.3	2.5×10^{-5}	2.0×10^{-5}
COW	24/25	52.5	35.2	114.7	1.4×10^{-5}	1.1×10^{-5}
POOL	25/25	151.7	78.1	239.4	4.1×10^{-5}	3.3×10^{-5}
Diffuse On-Site Sources						
B292	22/22	239.4	230.8	555.0	6.5×10^{-5}	5.1×10^{-5}
B331	26/26	688.2	598.5	1576.2	1.9×10^{-4}	1.5×10^{-4}
B514	25/26	122.7	67.0	214.2	3.3×10^{-5}	2.6×10^{-5}
B624	25/25	651.2	333.0	1380.1	1.8×10^{-4}	1.4×10^{-4}

a See Figures 4-1 and 4-2 for sample locations.
b Interquartile range.
c DCG = 3.7×10^{-3} Bq/mL (1×10^{-7} μCi/mL).
d 1 mSv = 100 mrem.
e Interquartile range not calculated. See Chapter 14, Quality Assurance.

Volume 2, Table 4-14 for biweekly data.) The highest median concentration was observed at location B331. This concentration was 6.9×10^{-7} Bq / mL (1.9×10^{-17} Ci / mL) and represents 0.0002 of the DCG. The highest biweekly tritium concentration, 1.6×10^{-6} Bq / mL (4.3×10^{-17} Ci / mL), was observed in June. If it were a yearly average, this concentration would represent 0.0004 of the DCG.

The B331 location is near the Tritium Facility (Building 331), which has ceased operations except for inventory reduction and cleanup activities. During this process, tritium-contaminated equipment slated for disposal is stored in a waste accumulation area and sent to Hazardous Waste Management facilities. During 1994, outgassing from such waste processing released an estimated 0.11×10^{12} Bq/L (3 Ci) of tritium to the atmosphere outside of Building 331.

The B292 location is near an underground retention tank that had previously leaked (see the section on Tank Systems Management in Chapter 2 of the 1993 Environmental Report for information regarding the B292 area).

The B624 location is situated in the Building 612 yard, which is dedicated to hazardous waste, radioactive waste, and mixed waste management activities. The yard consists of several areas where waste containers that are outgassing tritium are stored outdoors. The 1994 median concentrations at B292 and B624 are lower than the median concentrations in 1993.

Beryllium in Air

The median concentrations of airborne beryllium for the Livermore-site perimeter sampling locations are shown in **Table 4-7**. (See Volume 2, Table 4-15 for monthly data.) The highest value of 17.4 pg/m^3 occurred in the October composite at location COW. The median concentration is 0.00065 of the monthly ambient concentration limit of 10,000 pg/m^3 established by the Bay Area Air Quality Management District (BAAQMD).

Figure 4-7 is a plot of the median beryllium concentration at the Livermore-site perimeter from 1974 through 1994. The overall median concentration was calculated to be 0.002 of the ambient concentration guide. Unless there is a change in LLNL's operations, it is expected that the beryllium levels will remain unchanged.

Site 300

Airborne Radioactivity

Most gross alpha determinations at Site 300 were at or near the analytical limit of detection for the method. **Table 4-2** shows the monthly gross alpha and gross beta detection frequency, median, IQR, and maximum for sampling locations at Site 300. (See Volume 2, Table 4-3 for monthly data.) The monthly median gross alpha and gross beta concentrations are shown in **Figures 4-4** and **4-5**. The Site 300 gross beta results show a similar pattern to that of the Livermore-site results. Typical gross alpha activity is -1.0×10^{-11} Bq/mL (-2.7×10^{-22} Ci/mL).

Table 4-7. Beryllium on air filters (in pg/m^3), Livermore-site perimeter and Site 300, 1994.

Sampling Location[a]	Detection Frequency	Median	Interquartile Range	Maximum
Livermore Perimeter				
SALV	11/12	<4.9	<12.3	14.5
MESQ	12/12	4.9	7.3	14.9
CAFE	12/12	7.8	7.0	17.1
MET	12/12	4.6	7.9	12.6
VIS	12/12	3.7	5.8	10.8
COW	12/12	6.5	8.0	17.4
Site 300				
EOBS	11/12	4.8	<8.2	10.7
ECP	11/12	<4.6	<9.8	10.4
WCP	12/12	4.6	7.8	14.1
LIN	11/12	6.4	<11.1	18.4
GOLF	12/12	6.6	8.5	16.7
TFIR	12/12	9.2	10.5	24.1
NPS	11/12	<4.4	<8.8	10.5
WOBS	11/12	<5.0	<8.5	11.3
801E	12/12	6.5	14.7	27.7

[a] See Figures 4-1 and 4-3 for sampling locations.

Typical gross beta activity is 4.4×10^{-10} Bq / mL (1.2×10^{-20} Ci / mL). The primary sources of observed gross alpha and gross beta activity are naturally occurring radioisotopes of uranium and thorium and their decay products.

Table 4-3 lists the annual median activities, IQR, the fraction of the DCG, as well as the DCGs, of gamma-emitting radionuclides in samples from Site 300 and Tracy. (See Volume 2, Table 4-5 for monthly data.) All these radionuclides were measured at concentrations significantly below the DCGs. Of the nuclides tabulated, ^{7}Be, ^{40}K, ^{226}Ra, ^{228}Ra, and ^{228}Th are naturally occurring. The primary source of ^{137}Cs normally is long-term global fallout and resuspension.

Table 4-4 shows the median concentration of ^{239}Pu on air filter samples collected from Site 300. (See Volume 2, Table 4-9 for monthly data.) The highest concentration of ^{239}Pu was observed in the August composite at a level of 1.2×10^{-14} Bq / mL (3.2×10^{-25} Ci / mL, or 0.00002 of the DCG). **Table 4-5** shows the median concentration of ^{238}U, and ^{235}U and the ^{235}U / ^{238}U ratio on air samples from Site 300. (See Volume 2, Table 4-11 for monthly data.) The highest concentration of ^{238}U was observed in the October composite at a level of

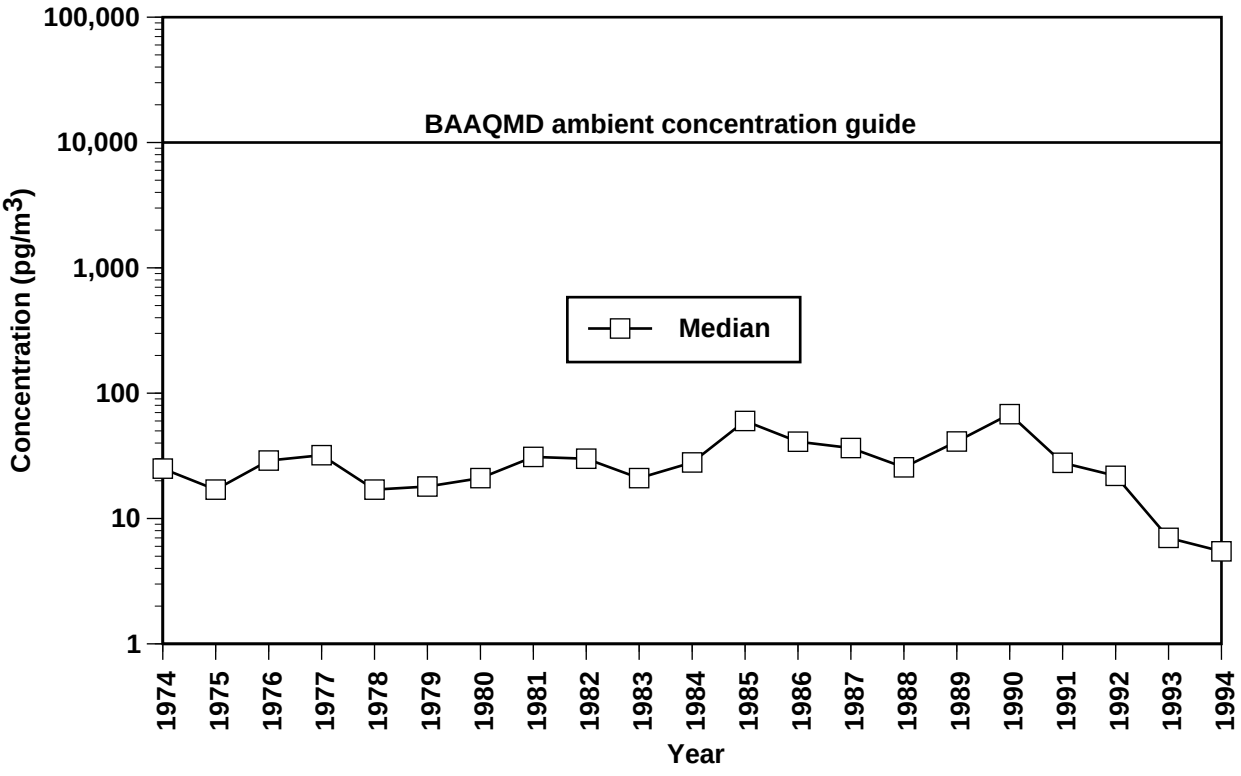

Figure 4-7. Median concentration of beryllium on air filters, Livermore-site perimeter, 1974 to 1994.

5.0×10^{-4} μg / m^3 (0.0017 of the DCG). The highest concentration of ^{235}U was observed in the October composite at a level of 1.5×10^{-6} μg / m^3 (0.00003 of the DCG). No other significant differences between locations or samples were noted. The overall levels were essentially the same as those reported in previous years.

The ratio of ^{235}U to ^{238}U can be used as an indicator of the source of the uranium. Both ^{235}U and ^{238}U occur naturally in the area, but only 0.7% of the naturally occurring uranium is ^{235}U, and the remainder is ^{238}U. Because Site 300 operations use depleted uranium that contains very little ^{235}U, it follows that if the ratio remains constant and near 0.7% (within the limit of sampling and analytical error), then the ^{238}U measured is from natural sources. The ^{235}U / ^{238}U ratio for October and December show statistically significant deviations from the natural ratio, indicating the presence of airborne depleted uranium from Site 300 operations. The measured concentrations of ^{238}U for 1994, however, are only 0.00016 of the DCG (DOE Order 5400.5). The ^{235}U / ^{238}U ratio for September indicates unrealistic levels of ^{235}U. A ratio of this magnitude would imply operations involving ^{235}U / ^{238}U ratios much higher than are actually used.

Because the September results were so unusual, reanalysis of the sample was requested. The reanalysis indicates naturally occurring concentrations of ^{235}U and ^{238}U.

Beryllium in Air

The detection frequency, median, IQR, and maximum concentrations of airborne beryllium for the Site 300 sampling locations are shown in **Table 4-7**. (See Volume 2, Table 4-16 for monthly data.) The highest beryllium concentration of 27.7 pg/m^3 occurred in August at location 801E. The concentration median is 0.0007 of the federal ambient concentration limit, which is 10,000 pg/m^3.

Environmental Impact

The environmental impacts from radioactive and nonradioactive effluents are described in this section.

Radioactive Effluents

Most of the tritium discharged to the atmosphere by LLNL in 1994 came from the Tritium Facility (Building 331). In 1994, operations there released a total of 5.1×10^{12} Bq (137 Ci) of tritium to the atmosphere. Of this, approximately 2.8×10^{12} Bq (76 Ci) were released as tritiated water (HTO). The remaining tritium was elemental tritium gas (HT). The highest single biweekly stack emission from the facility was 1.7×10^{11} Bq (4.7 Ci), of which 9.3×10^{10} Bq (2.5 Ci) was HT. This stack emission was measured between January 11–18, 1994. Sandia National Laboratories, California, released 3.4×10^{12} Bq (91 Ci) of HTO and 1.5×10^{12} Bq (4 Ci) of HT in 1994. Once released to the environment, the potential dose from tritium gas is approximately 25,000 times lower than a dose from a comparable release of tritiated water. Therefore, the tritiated hydrogen gas did not contribute significantly in calculations of the overall dose.

The potential for the release of radionuclides to the air from all discharge points from operations involving the use of radioactive materials is evaluated according to 40 CFR 61.93 of the NESHAPs regulations. This evaluation, performed on an annual basis, uses radionuclide inventories and monitoring data along with EPA-accepted release factors for operations and EPA-suggested reduction factors for emission control devices to estimate the potential release for each individual discharge point. Results have been published in LLNL NESHAPs 1994 Annual Report (Surano et al. 1995). An abbreviated-isotope summary of measured and calculated emissions for 1994 is presented in **Table 4-8**. The total estimated release from both point and diffuse sources for all isotopes used at LLNL was 5.5×10^{12} Bq (150 Ci). Tritium emissions account for 95% of the total estimated emissions. Emissions from point sources are 97% of the total emissions. A complete isotope listing of calculated emissions appears in Volume 2, Table 4-17.

Table 4-8. Calculated radioactive air emissions from the Livermore site for 1994.

Radionuclide[a]	Calculated Emissions[b] (Bq)	Radionuclide	Calculated Emissions[b] (Bq)
^{3}H (HTO)[c]	2.99×10^{12}	^{232}Th	4.81×10^{3}
^{238}U	1.16×10^{6}	^{244}Cm	2.20×10^{3}
^{241}Am	1.16×10^{5}	^{15}O	8.51×10^{10}
^{234}U	3.17×10^{5}	^{239}Pu	1.22×10^{3}
^{235}U	4.84×10^{4}	^{243}Cm	7.70×10^{2}
Gross alpha[d]	1.14×10^{4}	^{233}U	1.18×10^{3}
^{13}N	1.63×10^{11}	^{32}P	1.99×10^{7}
^{63}Ni	1.07×10^{9}	^{226}Ra	6.29×10^{3}
^{228}Th	9.62×10^{3}	^{3}H (HT)[c]	2.23×10^{12}
Total			**5.47×10^{12}**

a Radionuclides have been ordered by weighting the emissions according to the inhalation dose rate conversion factor for the isotope.

b Calculated emissions are estimates made according to NESHAPs 40 CFR 61.93 except those noted as measured. Values are considered to be conservative.

c Includes measured emissions.

d Gross alpha activity is reported in inventories where specific isotopic content is not determined.

Operations involving tritium at facilities other than the Tritium Facility had estimated releases totaling 0.18×10^{12} Bq (4.8 Ci) of HTO during 1994. The diffuse tritium sources at B292, B331, B514, and B624 have a localized effect; no elevated tritium concentrations were detected at the site perimeter or off site.

Estimated releases of the short-lived radionuclides ^{13}N and ^{15}O from Building 194 (the electron-positron linear accelerator) totaled 2.5×10^{11} Bq (6.7 Ci). Releases of ^{3}H and ^{13}N and ^{15}O radioactive effluents at LLNL during the 13-year period from 1981 through 1994 are shown in **Table 4-9**. The radioactive atmospheric emissions from these LLNL operations during 1994 are generally lower than previous years. (Note, in 1992 the Building 194 accelerator that generated ^{13}N and ^{15}O was not in operation.)

Analysis of air effluent samples for particulate emissions from facilities with monitoring (Buildings 175, 231, 251, 332, 419, 490, and 491) indicate air concentrations of alpha activity near or less than the MDL. Use of zero values for MDL data can be justified based on facility knowledge, use of tested multiple-stage HEPA filters, and isotopic analysis of filters. Isotopic analyses of the alpha activity of selected samples having values above the MDL have indicated the presence of activity from natural-occurring radon progeny such as polonium. Projecting MDL values for actual emissions, the estimated annual emissions of alpha activity associated with particles from the Livermore site yields 5.9×10^{5} Bq (1.6×10^{-5} Ci). The MDL-projected emissions have been substituted

Table 4-9. Radioactive airborne effluent releases from the Livermore site, 1981 through 1994.

Year	Airborne Effluents			
	^{3}H (GBq)[a]	^{3}H (Ci)[a]	^{13}N and ^{15}O (GBq)[b]	^{13}N and ^{15}O (Ci)[b]
1981	96,900	2,619	12,700	344
1982	74,520	2,014	21,600	584
1983	120,100	3,245	31,600	855
1984	272,100	7,354	3,000	81
1985	81,550	2,204	19,200	520
1986	46,400	1,254	4,180	113
1987	101,800	2,751	2,300	62
1988	147,400	3,983	1,100	30
1989	109,200	2,952	1,600	42
1990	47,430	1,282	1,800	48
1991	41,140	1,112	440	12
1992	6,550	177	0	0
1993	8,770	237	259	7
1994	5,070	137	248	7

a The tritium values reported are from Building 331 only. Additionally, an estimated 180 GBq were released during 1994 from other operations and diffuse sources.

b Estimated emissions from Building 194.

in screening calculations estimating dose; the total dose to the public attributable to LLNL operations is not significantly altered by these screening calculations.

The concentrations of radionuclides measured around Site 300 and in the City of Tracy were well below all standards and, except for uranium isotopes, reflect background or naturally occurring levels of these chemicals. The $^{235}U/^{238}U$ ratios in October and December are less than the ratio of naturally occurring concentrations of these isotopes, which suggests the presence of LLNL-induced depleted uranium in air samples from Site 300. These kinds of results can occur when tests using depleted uranium are conducted at Site 300. Estimated emissions from Site 300 operations that involve radioactive materials are calculated using inventories according to the NESHAPs regulations. Estimated releases of radionuclides for 1994 were 4.4×10^{10} Bq (1.2 Ci) of which 2.8×10^{9} Bq (7.6×10^{-2} Ci) were ^{238}U. Calculated emissions for Site 300 are presented in **Table 4-10**.

Table 4-10. Estimated radioactive air emissions from Site 300 for 1994.

Radionuclide	Quantity[a] (Bq)	Quantity[a] (Ci)
^{3}H	4.19×10^{10}	1.13
^{234}U	2.63×10^{8}	7.10×10^{-3}
^{235}U	3.59×10^{7}	9.70×10^{-4}
^{238}U	2.81×10^{9}	7.60×10^{-2}
Total	**4.50×10^{10}**	**1.22**

a Emissions are estimated according to NESHAPs 40 CFR 61.93.

All LLNL operations with measured and estimated radionuclide releases to the atmosphere and those with the potential to discharge radionuclides are evaluated for their potential impact to the public (see Chapter 12 on radiological dose assessment).

Nonradioactive Effluents

The concentrations of beryllium at both sites can be attributed to resuspension of surface soil containing naturally occurring beryllium. Local soils contain approximately 1 part per million (ppm) of beryllium, and the air of the Livermore area and Central Valley typically contains 10 to 100 $\mu g / m^3$ of particulates. Using a value of 50 $\mu g / m^3$ for an average dust load and 1 ppm for beryllium content of dust, an airborne beryllium concentration of 50 pg / m^3 can be calculated. The overall annual medians for the Livermore site and Site 300 are 5.5 pg / m^3 and 5.1 pg / m^3, respectively. These data are well below standards and do not indicate the presence of a threat to the environment or public health.

The estimated releases from exempt and permitted sources of air pollutants at the Livermore site can be compared to the most recent estimated 1994 daily release of air pollutants for the entire Bay Area. For example, the total emissions of oxides of nitrogen released in the Bay Area is approximately 444 metric tons per day compared to an estimate for LLNL releases of 0.065 metric tons per day (0.00015 of total Bay Area emissions). The BAAQMD estimate for reactive organic emissions is at 753 metric tons / day, versus Livermore site's estimated releases of 0.037 metric tons / day (0.00005 of total Bay Area emissions) in 1994.

Table 4-11 lists the estimated LLNL 1994 total releases for organic precursor and nonprecursor compounds, chlorofluorocarbons (an organic nonprecursor), and other LLNL airborne emissions.

Certain operations at Site 300 require permits from San Joaquin Valley Unified Air Pollution Control District. The total estimated air emissions during 1994 from operations (permitted and exempt air sources) at Site 300 are given in **Table 4-11**.

Table 4-11. Nonradioactive air emissions, Livermore site and Site 300, 1994.

Pollutant	Estimated Releases (metric tons/day)	
	Livermore Site	Site 300
Carbon monoxide	0.0109	0.00097
Chlorofluorocarbons	0.00477	0.000005
Organic compounds	0.0358	0.00155
Oxides of nitrogen	0.0646	0.00142
Oxides of sulfur	0.001	0.000045
Particulates	0.0092	0.00234